# Quantum Interference Mechanism of Cooperative Optical Phenomena in Extended Media

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In the quantum process of stimulated Raman scattering (SRS), a laser photon propagating in a resonance medium undergoes multifold conversions into a Stokes photon and back. The nontrivial "cooperative" behavior of the Stokes component of light transmitted through the medium is proven to be completely determined by the interference of scattering amplitudes in different sub-channels of the Stokes channel, which obviously combines all the sub-channels with an odd number of photon conversions. The theory of superfluorescence is then derived as the limiting case of the SRS theory.

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It has been typical to treat cooperative phenomena of resonance quantum optics in an extended medium, such as stimulated Raman scattering (SRS) [1] and superfluorescence (SF) [2], by first describing the initial growth of the Stokes pulse in SRS (or, respectively, the radiation emitted in a SF process) by linear quantum theory and then the succeeding evolution by the classical nonlinear equations. It is assumed that once the Stokes wave is initialized by the quantum vacuum fluctuations and the Stokes pulse has grown to a classical value, then the classical equations become valid. Both the classical Maxwell-Bloch (MB) equations describing SF [3] and a set of the classical equations describing SRS [4] were proven [5–11] to be integrable. That allows one to apply the inverse scattering method [12] to study the time evolution of the SF radiation and Stokes pulses within a medium.

In this Letter, we propose and discuss in detail a microscopic mechanism of cooperative optical phenomena based on studies of a quantum version of the standard classical model of SRS.

Propagating in a partially excited resonance medium, an incident laser photon undergoes multifold conversions into a Stokes photon and back. The number of these conversions defines a sub-channel of the scattering process. A scattering amplitude in the elastic (Rayleigh) channel is obviously given by a sum of scattering amplitudes in sub-channels with an even number of conversions. In this channel, an incident laser photon leaves a medium preserving the frequency. Correspondingly, a scattering amplitude in the inelastic (Stokes) channel is given by a sum of the scattering amplitudes in sub-channels with an odd number of conversions. Therefore, a photon leaves a medium as a Stokes one.

Analyzing explicit expressions for the scattering amplitudes, we show that a nontrivial "cooperative" behavior of the Stokes component of the radiation transmitted through a medium is completely determined by the interference of the scattering amplitudes in different sub-channels of the inelastic channel of scattering. The interference of the scattering amplitudes in different sub-channel of the elastic channel results correspondingly in the "cooperative" behavior of the Rayleigh component of transmitted light.

A solution of the quantum model of SRS shows that there is neither "linear quantum stage" nor "succeeding nonlinear classical evolution" of the Raman process. The quantum interference mechanism describes the "cooperative" character of SRS in a unified manner for all incident photons. Moreover, the scattering amplitudes do not depend on any physical parameters of an incident laser pulse, such as its amplitude, duration, etc. They are determined only by the quantum state of a medium. Scattering in a medium, even in the elastic channel, an incident laser photon changes the quantum state of a medium. This results in changing the scattering amplitudes for all successive photons, explaining thus the "cooperative" behavior of the transmitted light.

When the number of excitations of a medium grows, a number of possible inelastic sub-channels also grows, and the probability of scattering in the Stokes channel increases. However, if a number of excited atoms exceeds a half of the total number of atoms in a medium, further excitation results in a decrease in the number of inelastic sub-channels. Therefore, the probability of scattering in the Stokes channel begins to fall, and vanishes asymptotically in the limit of a completely inverted medium. The intensity of the Stokes radiation coming out of a medium has a pulse (or soliton-like) shape.

Finally, we show that a quantum model of SF in an extended medium (or a quantum version of the classical MB model) does not require a special consideration. Its solution is derived as the limiting case of a solution of the SRS problem with an appropriate initial condition.

The "cooperative" behavior of the radiation emitted by an initially excited system of two-level atoms is determined by quantum interference of amplitudes in different sub-channels of emission. In the SF case, the sub-channels of the

emission process are characterized by different numbers of possible reabsorption processes of a photon propagating in a partially excited medium.

Integrability of the quantum model of SRS allows one to derive an exact expression for the out-state of a scattering process [13]. Unfortunately, a complex algebra of operators involved in this expression does not admit an analytical computation of physical observables of the transmitted radiation. The problem of a computation of physical observables is equivalent to the problem of correlation functions in the theory of integrable quantum systems [14]. However, the analytical results suggest a simple scheme for numerical calculations and numerical simulations of the SRS process in terms of the six-vertex model of statistical mechanics [15].

The Hamiltonian of a quantum version of the standard SRS model can be written in the following form [13]:

$$H = -\int_{-\infty}^{\infty} dx \left\{ i\bar{\epsilon}_\sigma(x)\frac{\partial}{\partial x}\epsilon_\sigma(x) + J\sum_{a=1}^{M}\delta(x-x_a)\bar{\epsilon}_\sigma(x)\left[\sigma^+_{\sigma\mu}r^-_a + \sigma^-_{\sigma\mu}r^+_a\right]\epsilon_\mu(x)\right\}, \tag{1}$$

where the summation over repeated indexes is assumed. In Eq. (1), the operators of the "slow" envelopes of the laser (pump), $E_L(x)$, and Stokes, $E_S(x)$, components of light are combined into the isotopic spinor

$$\epsilon_\sigma = \begin{pmatrix} E_L \\ E_S \end{pmatrix}, \quad \bar{\epsilon}_\sigma = (E_L^\dagger, E_S^\dagger)$$

with the commutator $[\epsilon_\sigma(x), \bar{\epsilon}_\mu(y)] = \delta_{\sigma\mu}\delta(x-y)$, $\sigma, \mu = L, S$. To keep our terminology simple, we use hereafter the term "spin" for the field's subindex $\sigma$. Thus, the radiation field is described in terms of particles with spin up (a laser photon) and down (a Stokes photon).

To account for a possible saturation of a resonance transition of a medium, we treat the medium as a discrete set of $M$ two-level atoms with the coordinates $x_a$, $a = 1, \ldots, M$, along the light propagation axis - the $x$ axis. All the atoms are assumed to be positioned on an interval of size $l$, $0 < x_1 < \ldots < x_M < l$. As usual, they are described by the spin-$\frac{1}{2}$ operators $\mathbf{r}_a = (r^x_a, r^y_a, r^z_a)$, $r^\pm = (r^x \pm i r^y)/2$. The Pauli matrices $\sigma^\pm_{\sigma\mu}$ act in the spin space of the radiation. The first term of the model Hamiltonian describes a free propagation of the field components in the positive direction of the sample axis, while the second one represents a medium-field interaction with the coupling constant $J$, the two-level atoms playing the role of "spin impurities".

In the classical limit and in the limit of a continuous description of the medium, the equations of motion for the dynamical variables of the field plus medium system coincide with the equations of the classical SRS theory [1,2,4]. In the quantum approach, integrability of the SRS model is obvious, because the Hamiltonian (1) coincides with the Hamiltonian of the Kondo model with anisotropic exchange coupling whose integrability has been proven by Wiegmann [16].

The scattering of the $j$-th particle on the $a$-th impurity is described by the scattering matrix [16,13]

$$S_{ja} = \frac{1}{2}(1\otimes 1 + \sigma^z_j \otimes r^z_a) + \frac{b}{2}(1\otimes 1 - \sigma^z_j\otimes r^z_a) + c\,(\sigma^+_j\otimes r^-_a + \sigma^-_j\otimes r^+_a), \tag{2}$$

where $\otimes$ implies the tensor multiplication, and the scattering amplitudes $b = \cos J$ and $c = i\sin J$ correspond to the elastic (Rayleigh) and inelastic (Stokes) channels, respectively. For what follows, it is convenient to use the independence of the $S$-matrix of a particle momentum [17] and to formulate the SRS problem only in terms of spin variables of particles and impurities.

The initial state of the spontaneous SRS problem is then given by

$$|\mathrm{In}\rangle = |\bar{\Omega}_\mathrm{p}\rangle \otimes |\Omega_\mathrm{i}\rangle, \tag{3}$$

where the states of $N$ incident particles (all spins up) and $M$ impurities (all spins down) are defined as $\sigma^+_j|\bar{\Omega}_\mathrm{p}\rangle = 0$ and $r^-_a|\Omega_\mathrm{i}\rangle = 0$. It is convenient to represent the particle-impurity scattering matrix (2) as a $2\times 2$ matrix acting in the spin space of a particle, while its matrix elements contain the spin variables of an impurity,

$$S = \begin{bmatrix} \frac{1}{2}[(1+b) + (1-b)r^z_a] & c\,r^-_a \\ c\,r^+_a & \frac{1}{2}[(1+b) - (1-b)r^z_a] \end{bmatrix}.$$

The subsequent scattering of the $j$-th particle on the system of $M$ impurities is then described by the ordered scalar product of matrices $S_{ja}$,

$$L_j = S_{jM}\ldots S_{j1} = \begin{bmatrix} A & B \\ C & D \end{bmatrix},$$

where the operators $A, B, C$, and $D$ act in the spin space of impurities. Acting on the initial state (3), the monodromy matrix $L_j$ obviously reduces to the matrix

$$U_j = A + C \otimes \sigma_j^-,$$

where the first term corresponds to the elastic channel of photon scattering on the atomic system, while the second one represents the inelastic channel. In the latter, an incident laser photon is converted into a Stokes one, and simultaneously an excitation in the atomic system is created by the operator $C$. The exact final state of the spontaneous SRS problem is thus given by the ordered product of the matrices $U_j$,

$$|\mathrm{Out_N}\rangle = (A + C \otimes \sigma_N^-) \ldots (A + C \otimes \sigma_1^-)|\mathrm{In}\rangle. \tag{4}$$

Unlike the SRS problem in a pointlike geometry of the atomic system [13,18–21], the algebra of the elements of the monodromy matrix $L$ in an extended medium is not closed. Therefore, calculations of expectation values on the out-state (4) run into great mathematical troubles. Here, we compute the scattering amplitudes only for first two incident photons to clarify a microscopic quantum mechanism of the SRS phenomenon.

The first photon ($j = 1$) propagates in the unexcited atomic system and the explicit expression for the out-state is easily found to be

$$|\mathrm{Out}_1\rangle = b^M|\mathrm{In}\rangle + c\sum_{a=1}^{M} b^{(a-1)} r_a^+ |\Omega_i\rangle \otimes \sigma_1^- |\bar{\Omega}_p\rangle. \tag{5}$$

Eq. (5) has a clear physical meaning. The first term corresponds to the elastic channel with the scattering amplitude $\mathcal{A}_L^{(1)} = b^M$ and the probability $P_L^{(1)} = b^{2M}$. The second term represents the inelastic channel, in which the laser photon is converted into a Stokes photon, and simultaneously an excitation in the atomic system is created. The probability amplitude of finding the $a$-th atom in the excited state, $\phi_a = cb^{(a-1)}$, is naturally treated as the wave function of a one-particle medium excitation created by the transmitted photon.

In this expression, the term $c = i \sin J$ is the probability amplitude to excite the $a$-th atom, while $b^{(a-1)} = (\cos J)^{(a-1)}$ is the probability amplitude that the scattering on the preceding $a - 1$ atoms was elastic. Since $J \ll 1$, the wave function of the excitation takes an obvious exponential form

$$\phi(x) = iJ \exp{(-J^2 \rho x/2)}. \tag{6}$$

Here, aa atomic number $a$ is represented as $a = \rho x$, where $\rho = M/l$ is the linear density of the number of atoms, and $x$ is the atomic coordinate measured from the left edge of the medium.

Let us consider now the scattering process of the second incident laser photon ($j = 2$). This photon propagates in the medium whose quantum state has been prepared by the first photon. The probability of the elastic scattering of the second particle is obviously given by

$$P_L^{(2)} = \langle \mathrm{Out}_1 | AA | \mathrm{Out}_1 \rangle, \tag{7}$$

and we have

$$A|\mathrm{Out}_1\rangle = b^M b^M |\mathrm{In}\rangle + b^{(M-1)} c \left[\sum_{a=1}^{M} b^{(a-1)} r_a^+ + c^2 \sum_{a_1=1}^{M} \sum_{a_2=1}^{a_1-1} b^{(a_2-1)} r_{a_2}^+\right] |\Omega_i\rangle \otimes \sigma_1^- |\bar{\Omega}_p\rangle. \tag{8}$$

Here, the first term describes the elastic propagation of the particle in the unexcited medium, while the second and third terms correspond to the particle propagation in the medium excited by the first incident photon. The second term represents a sub-channel of the elastic channel of scattering in which a particle does not change the direction of its spin propagating within the medium as a laser photon, $1 \xrightarrow{L} M$. The third term represents the other possible sub-channel of the elastic scattering in which a particle changes the direction of its spin twice. Since one of the atoms, say $a_1$, is excited, a particle can change a spin direction first exciting an atom with a number $a_2 < a_1$, and then deexciting the atom $a_1$, $1 \xrightarrow{L} a_2 \xrightarrow{S} a_1 \xrightarrow{L} M$. Thus, on an interval between atoms $a_2$ and $a_1$ a particle propagates as a Stokes photon, but comes out of the medium as a laser photon.

Despite the factor $c^2$ being very small, the contribution of the third term to the probability $P_L^{(2)}$ is not small compared to the contribution of the second one. Moreover, omitting the third term, we immediately find that $P_L^{(2)} > P_L^{(1)}$, and hence the probability of the inelastic scattering falls with the growth of a photon number, $P_S^{(2)} < P_S^{(1)}$.

The existence of two sub-channels of scattering for the second incident particle plays a crucial role for the nontrivial behavior of the probabilities of scattering. The interference of the scattering amplitudes in two possible sub-channels completely determines the growth of the Stokes component. In the first sub-channel, the excited state of the atomic system is not changed, while in the second sub-channel an excitation of the atomic system created by the first incident photon is shifted to the left edge of the medium.

One obtains from Eqs. (7) and (8),

$$P_L^{(2)} = p^{2M} + \left[4 + p + \frac{M|c|^2(M|c|^2-4)}{1-p^M}\right] p^{(M-1)}(1-p^M), \tag{9}$$

where $p = b^2$, and hence, in the limit $J \ll 1$,

$$\frac{P_L^{(2)}}{P_L^{(1)}} = 1 - 2(MJ^2)^2 + \mathcal{O}[(MJ^2)^4]. \tag{10}$$

Thus, the interference of the scattering amplitudes in two sub-channels of the elastic channel of scattering drastically change the behavior of the probability $P_L^{(2)}$. Now, $P_L^{(2)} < P_L^{(1)}$, and hence, the probability of conversion of the second laser photon into the Stokes one, $P_S^{(2)} = 1 - P_L^{(2)}$, is bigger than this probability for the first transmitted photon, $P_S^{(1)} = 1 - P_L^{(1)}$, $P_S^{(2)} > P_S^{(1)}$.

The first derivative over time of the Stokes component intensity on the edge of an transmitted pulse, is easily found to be

$$\frac{d}{dt} I_S(t=0) = I_0 \frac{P_S^{(2)} - P_S^{(1)}}{P_S^{(1)}} \sim 2I_0(MJ^2)^2, \tag{11}$$

where $I_0$ is the intensity of an incident laser pulse. Thus, the intensity of the Stokes component at the initial moment of time grows with the velocity proportional to $M^2$; this is traditionally treated as a signature of the cooperative behavior of the SRS process.

Let us consider now an initial state of the scattering problem in which all incident particles are Stokes photons (all particle spins down), while all atoms are excited (all impurity spins up),

$$|\mathrm{In}\rangle = |\Omega_{\mathrm{p}}\rangle \otimes |\bar{\Omega}_{\mathrm{i}}\rangle, \tag{12}$$

where as $\sigma_j^- |\Omega_{\mathrm{p}}\rangle = 0$ and $r_a^+ |\bar{\Omega}_{\mathrm{i}}\rangle = 0$. Then, the transmitted radiation contains obviously both the Stokes and laser components. It is clear also that the scattering process results in a deexcitation of atomic system.

Let us consider the case of a single excited atom. The probability amplitude $A_{\mathrm{ex}}(N)$ to find the atom in the excited state after scattering $N$ incident laser photons is easily found from Eq. (2), $A_{\mathrm{ex}}(N) = (\cos J)^N$. Taking into account that $J \ll 1$, we obtain for a time evolution of the probability amplitude

$$A_{\mathrm{ex}}(t) = \left(1 - \frac{J^2}{2}\right)^N = \exp\left(-\frac{1}{2}\gamma t\right). \tag{13}$$

Here, $\gamma = J^2 \rho$, and the number of scattered photons at the moment of time $t$ is represented as $N = \rho t$, where $\rho$ is the linear density of incident photons.

As it should be expected, the excited atomic state decays exponentially with the decay constant $\gamma$ to the ground state. If

$$\rho \to \infty,\ J \to 0,\ \text{but}\ J^2 \rho = \gamma = \text{constant}, \tag{14}$$

as is already assumed in Eq. (13), an incident field is obviously eliminated from consideration, and the decay can be treated as spontaneous one, in which a single "Stokes" photon is emitted. In this limit, we can replace $JE_L \to \sqrt{\gamma}$ and $E_S(x) \to E(x)$ to reduce the Hamiltonian of the SRS problem (1) to the standard Hamiltonian of the quantum Maxwell-Bloch model,

$$H = -i \int dx E^\dagger(x) \frac{\partial}{\partial x} E(x) - \sqrt{\gamma} \sum_a \int dx \left[E^\dagger(x) r_a^- + r_a^+ E(x)\right] \delta(x - x_a). \tag{15}$$

An equivalence of these two models in the limit (14) has been used [22] to study the SRS problem in the framework of Maxwell-Bloch equation. It is clear that a solution of the more general SRS problem can be reduced to the SF problem only under some special conditions [22]. However, a solution of the SF problem, including statistical properties of emitted photons [23], is exactly derived from a solution of the SRS problem in the limit (14).

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